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Thermodynamic modeling of acid gas removal from natural gas using the Extended UNIQUAC model

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Abstract

Thermodynamics of natural gas sweetening process needs to be known for proper design of natural gas treating plants. Absorption with aqueous N-Methyldiethanolamine is currently the most commonly used process for removal of acid gas (CO_2 and H_2S) impurities from natural gas. Model parameters for the Extended UNIQUAC model have already been determined by the same authors to calculate single acid gas solubility in aqueous MDEA. In this study, the model is further extended to estimate solubility of CO_2 and H_2S and their mixture in aqueous MDEA at high pressures with methane as a makeup gas.

Keywords: Thermodynamics, Extended UNIQUAC, High pressure, MDEA, Acid gas, Natural gas

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1. Introduction

Natural gas is often recovered from the well at high pressures, around 5 to 10 MPa. Natural gas is usually contaminated with acid gases (CO_2 and H_2S).[1] Natural gas sweetening is carried out in order to remove these gases.

In order to avoid extra costs of pressurizing for liquefaction, the sweetening process must be carried out at high pressures, hence, without a pressure drop.[2]

Stripping acid gases from natural gas is commonly done by use of aqueous alkanolamines in an absorber-desorber system.[1] In natural gas treatment process, the typical absorber pressure is around 5 to 10 MPa, but the stripper pressure is between 0.1 and 0.3 MPa. In the absorber, mixtures of acid gas-hydrocarbons, mainly methane, are contacted counter currently with aqueous amines while in the stripper, mixtures of acid gas-amine-water are present as methane and other hydrocarbons are already separated. During the years different types of amines have been developed and used for specific gas treating applications.[3] Among amines, Methyldiethanolamine (MDEA) is usually preferred for full absorption of H_2S and partial absorption of CO_2.[3] MDEA can reduce the amount of H_2S and CO_2 to approximately 4 ppmv and 3 mol %, respectively.[3] Representation of the thermodynamic behavior of acid gas removal process form natural gas at operational conditions is important for proper design of natural gas treating plants. The objective of this study is to extend the Extended UNIQUAC model developed earlier by Sadegh et al.[4,5] to describe single and mixed acid gas solubility in aqueous MDEA in presence of methane as a makeup gas (at high pressures).

2. Model description

In this study the Extended UNIQUAC model[6] is developed to describe thermodynamic behavior of the H_2S-CO_2-CH_4-MDEA-H_2O system. The Extended UNIQUAC model is used to calculate activity

coefficients in the liquid phase and SRK[7] equation of state is applied for calculation of fugacities in the vapor phase. In the Extended UNIQUAC model the extended Debye – Hückel term is added to the original UNIQUAC[8,9] model in order to allow the model to be used for electrolyte solutions. The model structure has already been explained by Sadegh et al.[4,5]

3. Equilibrium calculations

For rigorous modeling of acid gas-alkanolamine system, both physical and chemical equilibrium have to be incorporated in the model.

Physical equilibrium

Molecular species distribute between liquid and vapor phase. The following vapor-liquid equilibria take place in the studied system.

$$CO_2\ (aq) \leftrightarrow\ CO_2\ (g) \quad (1)$$

$$H_2S\ (aq) \leftrightarrow\ H_2S\ (g) \quad (2)$$

$$CH_4\ (aq) \leftrightarrow\ CH_4\ (g) \quad (3)$$

$$H_2O\ (l) \leftrightarrow\ H_2O\ (g) \quad (4)$$

$$MDEA\ (aq) \leftrightarrow MDEA\ (g) \quad (5)$$

Chemical equilibrium

Acid gas-alkanolamine system is a reactive system, many reactions occur in the system. The following reactions are considered in this study.

Ionization of water: $H_2O\ (l) \leftrightarrow OH^-(aq) + H^+(aq)$ (6)

Protonation of MDEA: $MDEA\ (aq) + H_2O\ (l) \leftrightarrow MDEAH^+(aq) + OH^-(aq)$ (7)

Dissociation of carbon dioxide: $CO_2(aq) + OH^-(aq) \leftrightarrow HCO_3^-(aq)$ (8)

Dissociation of bicarbonate ion: $HCO_3^-(aq) + OH^-(aq) \leftrightarrow CO_3^{2-}(aq) + H_2O\ (l)$ (9)

Hydrogen sulfide dissociation: $H_2S\ (aq) \leftrightarrow H^+(aq) + HS^-(aq)$ (10)

Bisulfide ion dissociation reaction ($HS^-\ (aq) \leftrightarrow H^+(aq) + S^{2-}(aq)$) is excluded for modeling work of this study because the amount of sulfide ion is very small and negligible.[5]

4. Evaluation of model parameters

All adjustable model parameters belong to the UNIQUAC term and there is no adjustable parameter in $Debye - Hückel$ term and SRK EoS. The adjustable model parameters are UNIQUAC volume parameter (r_i), surface area parameter (q_i) and binary interaction parameters between species *"i"* and *"j"*, u_{ij}. The binary interaction parameter is symmetric ($u_{ij} = u_{ji}$) and temperature dependent.

$$u_{ij} = u_{ij}^0 + u_{ij}^T (T - 298.15) \quad (11)$$

Values of u_{ij}^0 and u_{ij}^T are adjusted to the experimental data. The values of u_{ij}^0 and u_{ij}^T for the binary interaction parameter between species that have a low possibility of coexistence in the solution has been set to 10^{10} and 0, respectively. The assigned values keep the ineffective parameters away from the regression process.

Evaluated experimental vapor-liquid equilibrium data (total pressure and acid gas partial pressure) were regressed in order to find the optimum values of model adjustable parameters. The model parameters were optimized to obtain the minimum of the objective function (S):

$$S = \sum_{VLEdata} \left[\frac{P_{calc} - P_{exp}}{w\,(P_{exp} + 0.01\ bar)} \right]^2 \qquad (12)$$

In equation (12), *"calc"* and *"exp"* are calculated and experimental values, respectively. *P* is either the solution total pressure (bar) or acid gas partial pressure (bar). *w* is the weight given to vapor-liquid equilibria data and it set to 0.05. This value was chosen based on the accuracy of the regressed data. 0.01 bar is added to the denominator of the VLE term in order to avoid giving too much weight to low pressure data.

5. Model parameters

5.1. Acid gas-CH_4-MDEA-H_2O systems

The modeling of the CO_2-CH_4-MDEA-H_2O and the H_2S-CH_4-MDEA-H_2O quaternary systems was started by determining model parameters for the CH_4-H_2O binary subsystem.

Methane dissolves only physically in the water. Vapor-liquid equilibrium should be taken into account to obtain methane physical solubility in water. To model the CH_4-H_2O system, the binary parameter for interaction between CH_4 and H_2O has been fitted to the binary VLE data for CH_4-H_2O system. The volume and surface area parameters, r and q, for methane and water were respectively taken from Addicks et al.[10] and Abrams and Prausnitz[8]. The values of the adjusted UNIQUAC parameters required for modeling CH_4-H_2O mixture are shown in Table 1. The determined parameter sets for the CH_4-H_2O system were then combined with the parameter sets of ternary acid gas-MDEA-H_2O from Sadegh et

al.[4,5] work to form a model for quaternary acid gas-CH_4-MDEA-H_2O system. The behavior of the quaternary acid gas-CH_4-MDEA-H_2O system can be well predicted by the combined model and there is no need to adjust any additional parameter.

5.2. Mixed acid gas-CH_4-MDEA-H_2O system

Mixed acid gas system can be modeled based on the parameters for single acid gases. When CO_2 and H_2S are both present, the additional HCO_3^--HS^- and CO_3^{2-}-HS^- interaction parameters are required compared to single acid gas systems. The H_2S-CO_2 interaction parameter was found to have no effect on modeling the mixed acid gas system. This interaction parameter was set to a large value which indicates no contribution to the excess Gibbs energy function. The parameters for the interactions HCO_3^--HS^- and CO_3^{2-}-HS^- were fitted to the mixed acid gas data. The adjusted values are presented in Table 1.

Table 1. $u_{ij}^0 = u_{ji}^0$ and $u_{ij}^T = u_{ji}^T$ parameters required for calculating UNIQUAC energy interaction parameters

Pair	$u_{ij}^0 = u_{ji}^0$	$u_{ij}^T = u_{ji}^T$
CH_4-H_2O	44.16483	1.4836
HCO_3^--HS^-	494.76	0.2494
CO_3^{2-}-HS^-	262.13	3.685

6. Results and discussion

6.1. Acid gas-CH_4-MDEA-H_2O systems

In this study, the phase behavior of the H_2S-CO_2-CH_4-MDEA-H_2O system has been correlated by combining the models for the constituent subsystems and determining interaction parameters for the

additional interactions. Before modeling acid gas-CH_4-MDEA-H_2O mixtures, model parameters for the CH_4-H_2O binary subsystem have to be established.

Regression results for the binary CH_4-H_2O system

The value of the CH_4-H_2O binary interaction parameter required for modeling CH_4-H_2O system was adjusted to 31 total pressure data of CH_4-H_2O mixtures. Table 2 shows regression results for the total pressure of CH_4-H_2O system.

Table 2. Overview on binary CH_4-H_2O data used for parameter optimization and regression results

CH_4 Concentration, Molality	T, K	P_{Total}, kPa	Data Type	Reference	Number of Data Points	AARD[a]%
0.02 to 0.09	298.15, 323.15	3000 to 8000	VLE	Yokoyama et al.[11]	6	1.9
0.03 to 0.22	283.15, 293.15, 303.15	2000 to 40030	VLE	Wang et al.[12]	17	7.6
0.01 to 0.09	298.75, 314.15	993 to 9981	VLE	Awan et al.[13]	8	5

[a] Average Absolute Relative Deviation: AARD = $\frac{\sum_{i=1}^{N} \frac{(|z_{i,exp} - z_{i,calc}|)}{z_{i,exp}}}{N}$

Results of fit for total pressure of CH_4-H_2O binary system at 298.15, 314.15 and 323.15 K for data of Yokoyama et al.[11] and Awan et al.[13] are shown in Figure 1. Model calculations against experimental data of Yokoyama et al.[11] and Awan et al.[13] are plotted in the figure. Uncertainty of the measured pressure data is estimated to be within ± 0.003 MPa (3 kPa) and ± 0.1 mmHg (0.013 kPa) for Awan et al.[13] and Yokoyama et al.[11] data, respectively. [11,13]

Since the error bars for the experimental points are very small amount compared to the measured points, they cannot be seen in the figure.

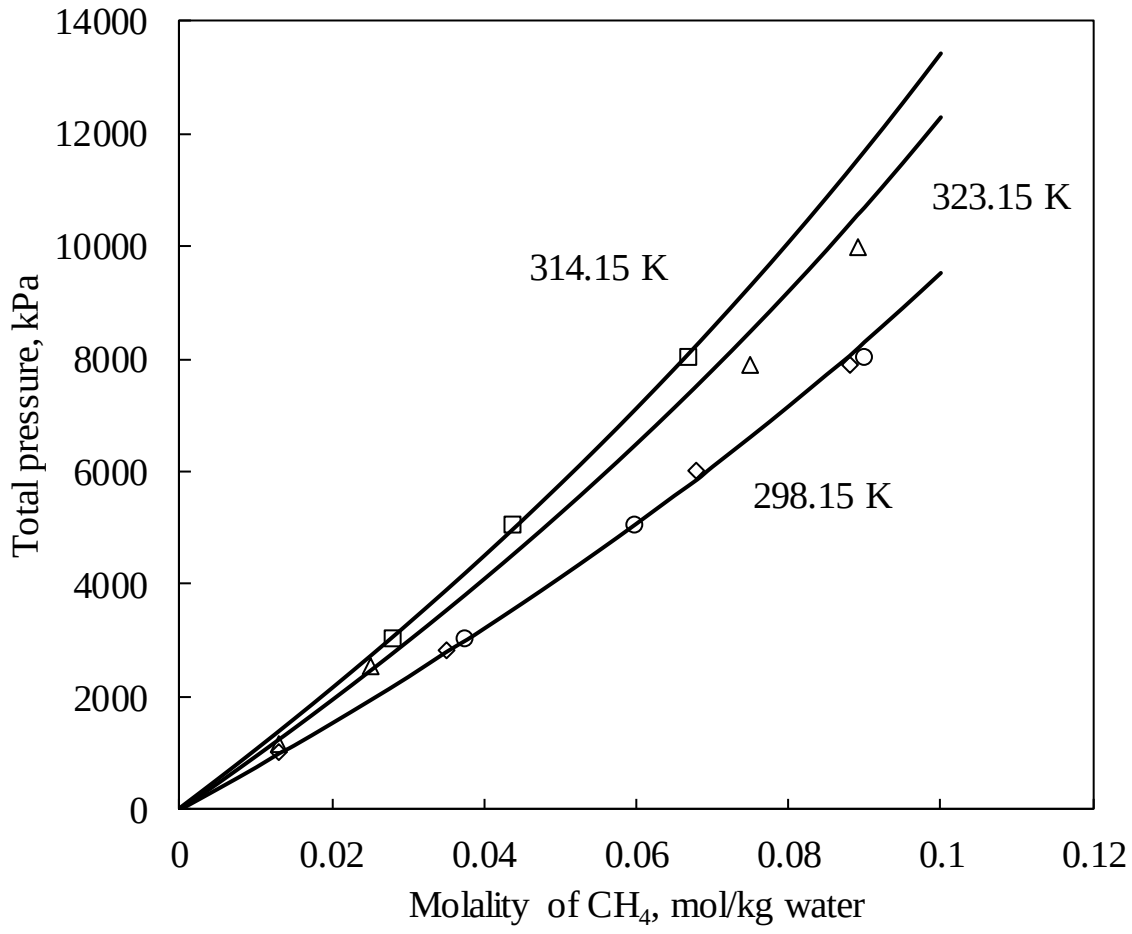

Figure 1. Comparison between calculated (lines) and experimental total pressure of CH_4-H_2O solution. ○ **(T = 298.15 K),** □ **(T = 323.15 K), Yokoyama et al.[11];** ◇ **(T =298.15 K),** △ **(T = 314.15 K), Awan et al.[13]**

As it can be seen from figure 1, there is a good agreement between calculated values and experimental data from Yokoyama et al.[11], While there is a larger deviation between calculated results and experimental values of Awan et al.[13].

Overall, the developed model represents total pressure of CH_4-H_2O system within AARD % of 4.8.

Prediction results for CO_2-CH_4-MDEA-H_2O and H_2S-CH_4-MDEA-H_2O systems

The previously determined parameter sets for single gases in aqueous amine solutions from Sadegh et al.[4,5] were combined with the parameters determined in this work for CH_4-H_2O subsystem to create a predictive tool for representing the behavior of acid gas-CH_4-MDEA-H_2O mixtures. Table 3 lists data sources which were used to validate the model predictions for the quaternary acid gas-CH_4-MDEA-H_2O systems.

Table 3. Prediction results for acid gas partial pressure for acid gas-CH_4-MDEA-H_2O mixture

MDEA Concentration, wt %	T, K	Total Pressure, kPa	Acid Gas Partial Pressure, kPa	Data Type	Reference	Number of Data Points	AARD[a]%
50	323.15	499 to 700	3 to 278 (P_{H_2S})	VLE (P_{H_2S})	Dicko et al.[14]	5	15
34.99, 49.99	283.15, 298.15	690, 3450, 6900	0.1 to 18 (P_{H_2S})	VLE (P_{H_2S})	Huttenhuis et al.[15]	30	15
50	313.15	350	0.2 to 4.9 (P_{H_2S})	VLE (P_{H_2S})	Ter Maat et al.[16]	7	5.8
30, 50	313.15, 353.15	10000, 15000, 20000	11 to 5066 (P_{CO_2})	VLE (P_{CO_2})	Addicks et al.[10]	31	21
50	323.15	1268 to 1558	6 to 434 (P_{CO_2})	VLE (P_{CO_2})	Dicko et al.[14]	5	30

[a] As described above (See Table 2)

Figure 2 shows model predictions against experimental data of Ter Maat et al.[16] for H_2S partial pressure for H_2S-CH_4-MDEA-H_2O system at total pressure of 350 kPa. The overall accuracy of the

measured partial pressure by Ter Maat et al.[16] is estimated to be ± 5%.[16] The error bars for the experimental points are shown in the figure.

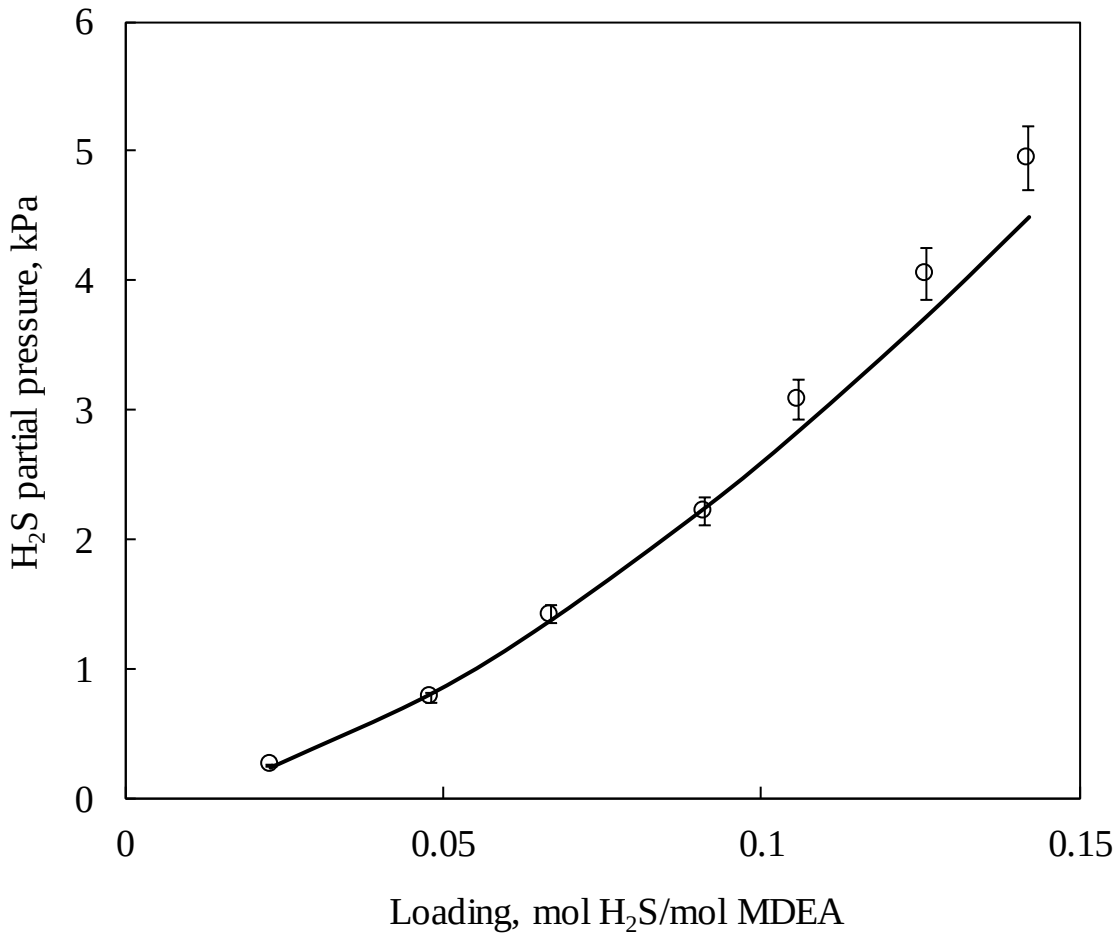

Figure 2. Comparison between model predictions (line) and experimental data for H_2S solubility in 50 wt % aqueous MDEA at 313.15 K and total pressure of 350 kPa with methane as a makeup gas. ○, Ter Maat et al.[16]

Figure 3 depicts model predictions against experimental data of Huttenhuis et al.[15] for H_2S solubility in aqueous MDEA in presence of methane as a makeup gas at total pressures of 3450 kPa. It is noted that

accuracy of the experimental measurements was not mentioned in Huttenhuis et al.[15] paper or published paper.

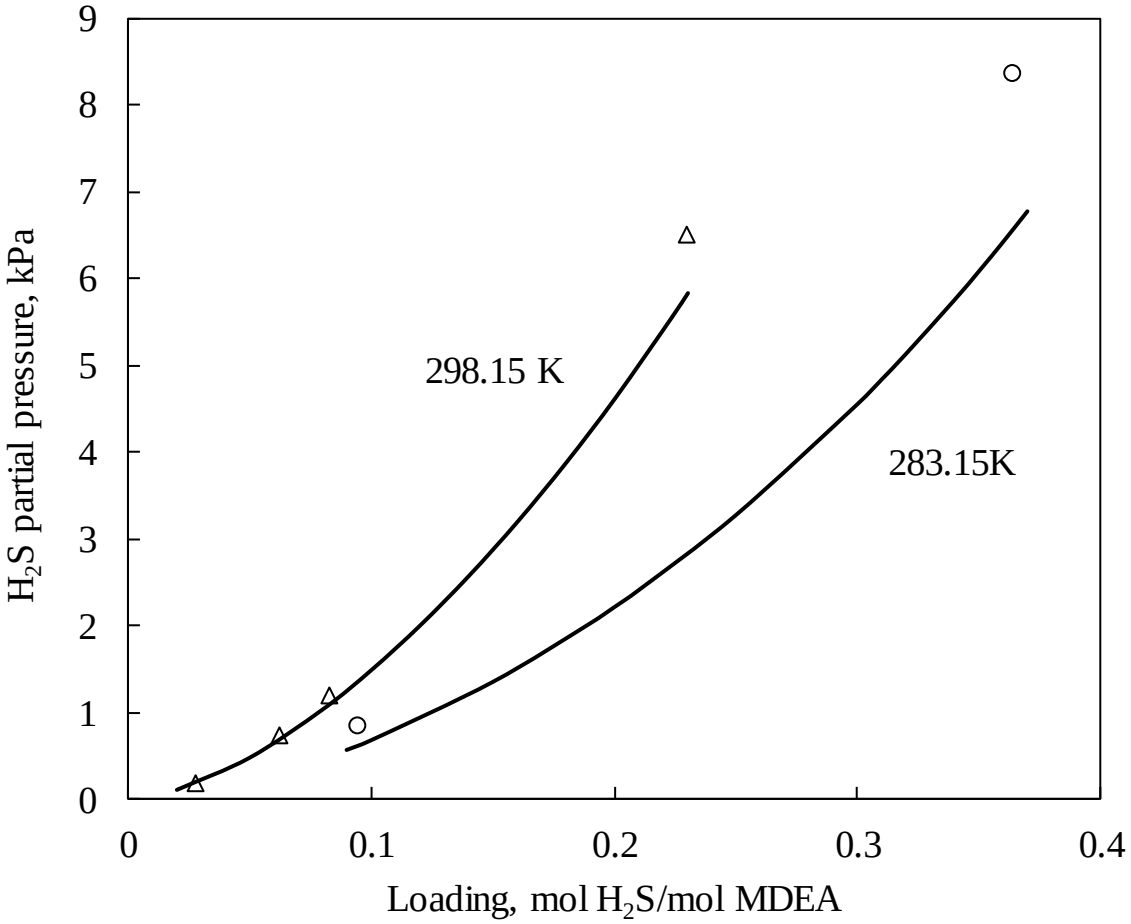

Figure 3. Comparison between predicted (lines) and experimental values for H_2S solubility in 49.99 wt % aqueous MDEA at 10 and 25 ˚C and total pressure of 3450 kPa with methane as a makeup gas. ○, (T=283.15 K), ∆, (T=298.15 K), Huttenhuis et al.[15]

In Figure 4, predicted and experimental measurements of Addicks et al.[10] for CO_2 partial pressure for CO_2-CH_4-MDEA-H_2O system is plotted against loading at total pressure of 10000 kPa. The pressure

measurements were performed with a high pressure sensor up to 1000 bar (100000 kPa) with accuracy of ± 0.1% of full scale.[10] So accuracy of pressure data by Addicks et al.[10] is estimated to be 100 kPa. Error bars for pressure measurements are shown in the figure.

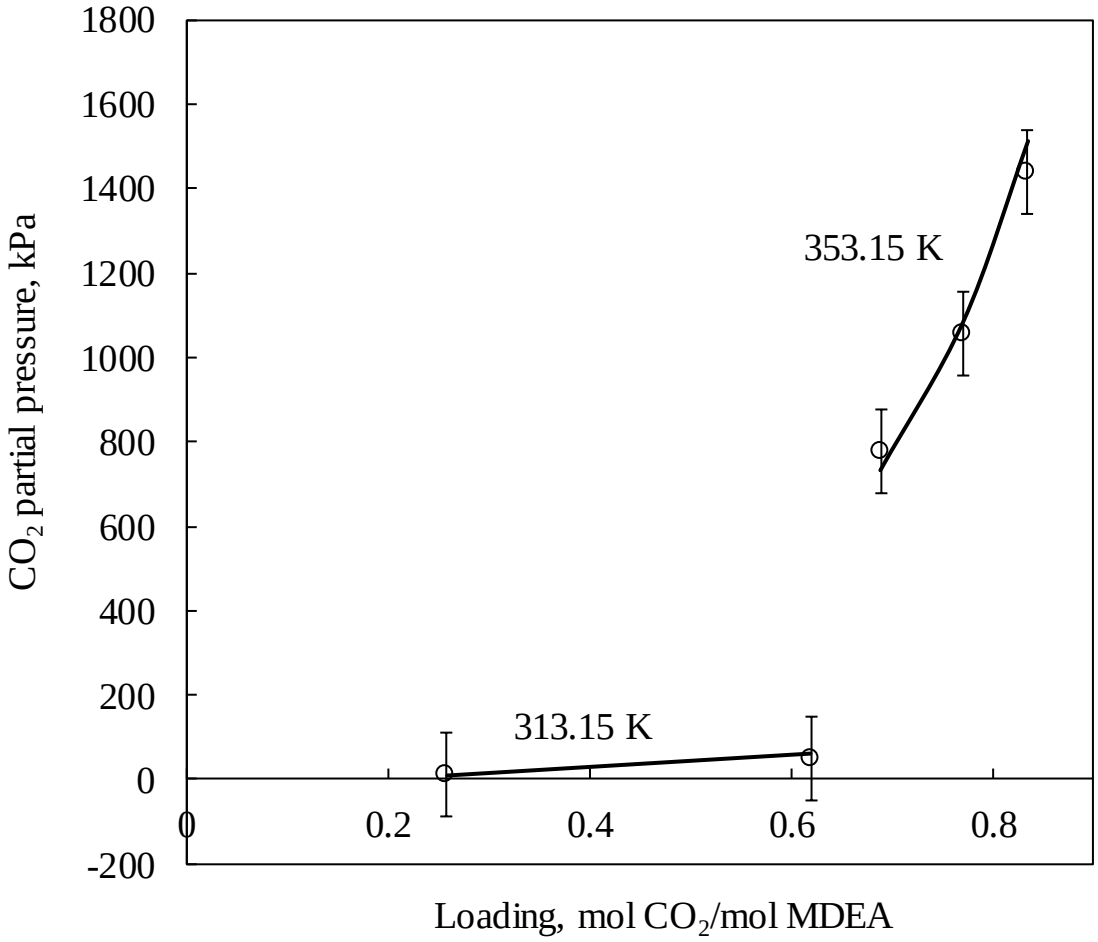

Figure 4. Comparison between model predictions (lines) and experimental data for CO_2 solubility in 30 wt % aqueous MDEA at 313.15 K and 353.15 K and total pressure of 10000 kPa with methane as a makeup gas. ○, Addicks et al.[10]

As it can be seen from the above figures, predicted values for acid gas solubility in aqueous MDEA with methane as a makeup gas properly agree with the experimental data which were not used for regression. Totally, the model predicts H_2S and CO_2 solubility in aqueous MDEA with CH_4 as a makeup gas within AARD % of 11 and 25, respectively.

6.2. Mixed acid gas-CH_4-MDEA-H_2O system

To model the behavior of the H_2S-CO_2-CH_4-MDEA-H_2O system, the parameters of single acid gas systems were combined and additional required interaction parameters were identified. The required additional parameters were regressed to data of the H_2S-CO_2-CH_4-MDEA-H_2O system. The experimental data upon which model parameters were optimized are presented in Table 4 together with the regression results.

Table 4. Regression results for acid gas partial pressure for H_2S-CO_2-CH_4-MDEA-H_2O mixture

MDEA Concentration, wt %	T, K	Total Pressure, kPa	H_2S Partial Pressure, kPa	CO_2 Partial Pressure, kPa	Reference	Number of Data Points	AARD[a]%	
							H_2S	CO_2
34.90, 50.02	283.15, 298.15	100 to 6900	0.12 to 35.20	0.08 to 14.87	Ter Maat et al.[16]	72	23	31

[a] As described above (See Table 2)

Figures 5 and 6 are parity plots for CO_2 and H_2S partial pressure over mixtures of H_2S-CO_2-CH_4-MDEA-H_2O, respectively. The figures respectively represent model calculations for CO_2 and H_2S partial pressure for the H_2S-CO_2-CH_4-MDEA-H_2O system against regressed experimental data of Ter Maat et al.[16] The accuracy for acid gas partial pressure data of Ter Maat et al.[16] is estimated to be ± 5%.[16] The accuracy limit for the experimental data is shown by the dash round dot lines in figures 5 and 6.

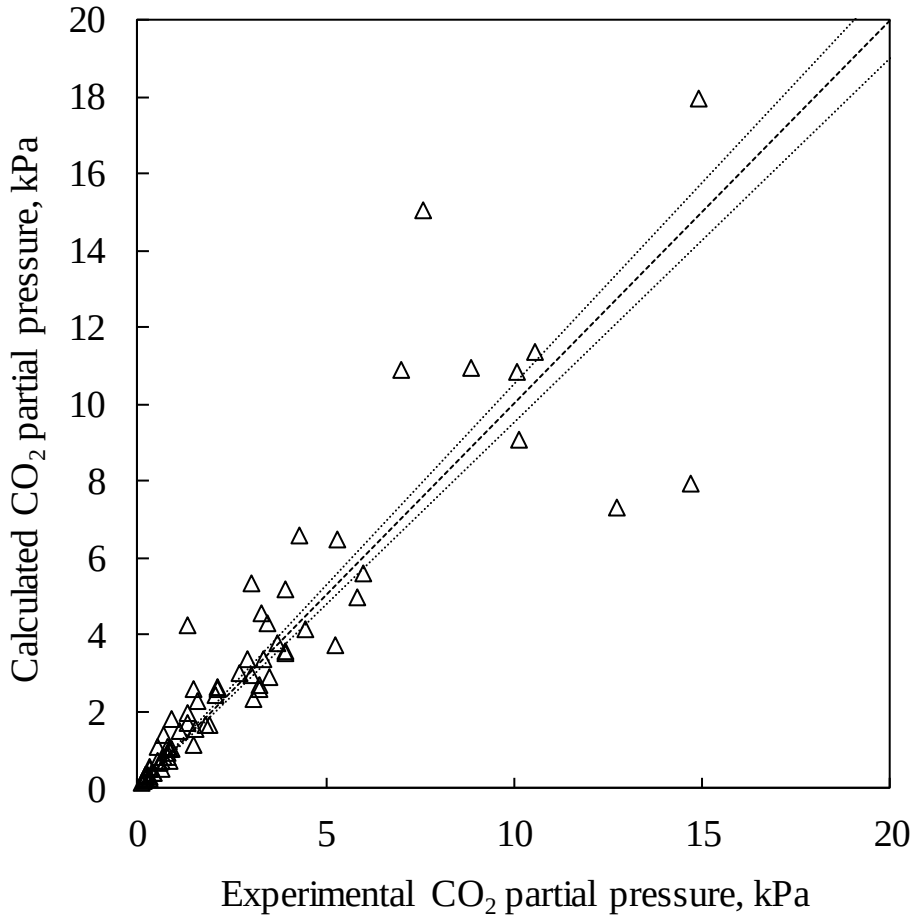

Figure 5. Parity plot for CO_2 partial pressure over H_2S-CO_2-CH_4-MDEA-H_2O mixture. Δ, Ter Maat et al.[16]

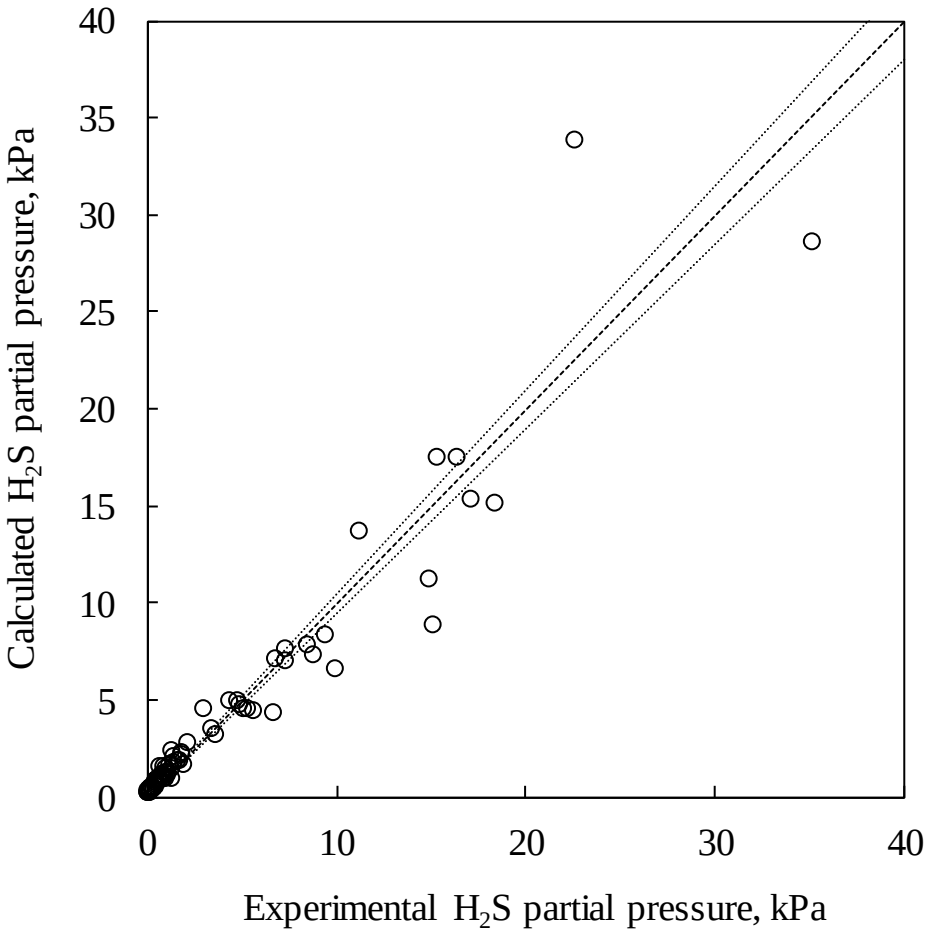

Figure 6. Parity plot for H_2S partial pressure over H_2S-CO_2-CH_4-MDEA-H_2O mixture. ○, Ter Maat et al.[16]

As it can be seen from figures 5 and 6, for CO_2 partial pressures below 5 kPa and H_2S partial pressure below 10 kPa most of the calculated partial pressures lies between the accuracy limit of the experimental data. Indicating a good agreement between measured and calculated CO_2 and H_2S partial pressures at low pressures.. However, for CO_2 partial pressures higher than 5 kPa and H_2S partial pressure over10 kPa , there is a larger difference between model calculations and experimental data. At these pressures calculated values are not within experimental accuracy. This large difference between

calculated and measured values at higher pressures can be interpreted as the results of either error in experimental measurements or model deficiency at higher pressures. The accuracy of the higher pressure measurements of Ter Maat et al.[16] cannot be assured. Because at the time of modelling the only available source of data was Ter Maat et al.[16] and it was not possible to check the reliability of the reported data by comparing to other sources. If the reported accuracy by Ter Maat et al.[16] is valid over the whole range of the pressure measurements and by considering all of the reported data accurate are accurate, then the large deviation at higher pressure is because of not well-tuned model parameters. If this is the case, that can be because model parameters for mixed acid gas system were tuned only to Ter Maat et al.[16] data, as this was the only available source. And in this source, there are smaller number of data at higher pressures compare to low pressures. So model parameters were tuned to limited number of data at higher pressures.

The model represents H_2S and CO_2 partial pressures for H_2S-CO_2-CH_4-MDEA-H_2O system within AARD % of 23 and 31, respectively

7. Conclusion

Accurate representation of single and mixed acid gas solubility in aqueous MDEA at high pressure and in the presence of methane is important for the proper design of natural gas sweetening process. In this study, Extended UNIQUAC model parameters have been determined to enable the model to describe thermodynamic properties of the CO_2-CH_4-MDEA-H_2O, H_2S-CH_4-MDEA-H_2O and H_2S-CO_2-CH_4-MDEA-H_2O systems. Modeling of acid gas-CH_4-MDEA-H_2O system began with creating a model for the CH_4-H_2O subsystem. The acid gas-CH_4-MDEA-H_2O then was modeled by incorporating CH_4-H_2O parameters and already determined parameters for acid gas-MDEA-H_2O system into one single set of parameters and with no need to add any additional adjustable parameter. The H_2S-CO_2-CH_4-MDEA-H_2O system was modeled by combining parameters of CO_2-CH_4-MDEA-H_2O and H_2S-CH_4-MDEA-

H_2O systems and adjusting additional required parameters related to the mixed acid gas system to mixed acid gas data.

Acknowledgement

We would sincerely appreciate Statoil ASA for financial support of the project.

Nomenclature

Abbreviations

VLE: Vapor-Liquid Equilibrium

AARD: Average Absolute Relative Deviation

ppmv: parts per million by volume

CO_2: Carbon dioxide

H_2S: Hydrogen sulfide

CH_4: Methane

H_2O: Water

MDEA: N-Methyldiethanolamine

UNIQUAC: UNIversal QUAsi Chemical thermodynamic model

T: Temperature

P: Pressure

Calc.: Calculated

Exp.: Experimental

Symbols

P_{Total}: Total Pressure

P_{CO_2}: CO_2 partial pressure

P_{H_2S}: H_2S partial pressure

r_i: UNIQUAC volume parameter

q_i: UNIQUAC surface area parameter

u_{ij}: UNIQUAC binary interaction parameter

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